Picosecond Dynamics Probed by X-ray Diffraction

Pump-probe time-resolved x-ray diffraction of allowed and nearly forbidden reflections in InSb is used to follow the propagation of a coherent acoustic pulse generated by ultrafast laser excitation. The surface and bulk components of the strain can be measured simultaneously because of the large x-ray penetration depth.

X-ray diffraction has long been used as a tool for structural determination. Its usefulness is due primarily to the short wavelength of x-ray radiation, which is comparable to the interatomic distances in solids. Crystals form periodic arrays of atoms or molecules (the lattice), which diffract x-rays when they are of a proper wavelength and direction of incidence, determined by the microscopic placement of the individual atoms. X-ray cyrstallographers work backwards from these often complicated diffraction patterns to determine the underlying structure of the material. Scientists at the Center for Real-Time X-ray Studies at the Advanced Photon Source (as part of the Michigan/Howard/Lucent Technologies, Bell Labs Collaborative Access Team) are using the x-rays emitted from the synchrotron to study the structural dynamics of materials on an ultrafast time scale. They are using a technique called pump-probe time-resolved x-ray diffraction, in which a high-power laser (see Fig. 1) acts as a pump to initiate structural changes in a material, after which a short-duration x-ray pulse probes the structure of the evolving lattice. By repetitively exciting the sample and varying both the time delay of the probe pulse and the crystal orientation, the scientists gradually build up a picture of the time evolution of the lattice. Much as a camera can stop action only as fast as the shutter speed, the x-rays are only able to stop action on a time limited by the pulse duration. In the synchrotron, the x-rays are emitted in a series of short pulses about 100 picoseconds each in duration (one ten-billionth of a second).

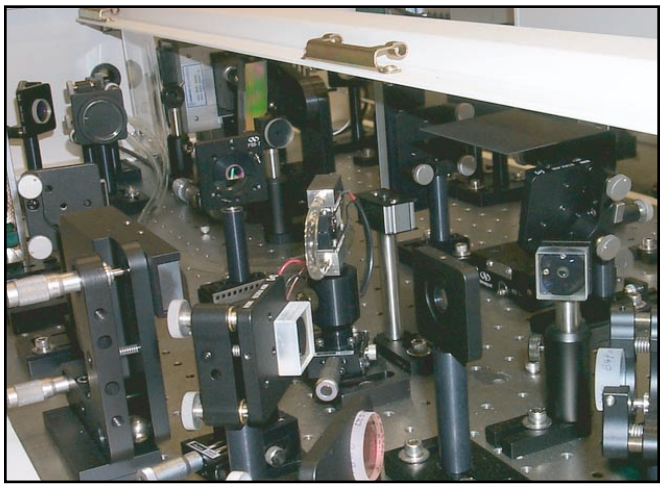

FIG. 1. Photograph showing the ultrafast Ti:sapphire laser amplifier.

These pulses are sufficiently short to study a variety of atomic and molecular dynamics. The power of this technique was recently demonstrated in an experiment on ultrafast strain generation and propagation in semiconductors; the results are published in *Physical Review Letters* [1]. Figure 2 shows the time-resolved diffraction pattern obtained for an InSb crystal that has been rapidly heated by the absorption of a laser pulse of only a few tens of femtoseconds in duration (approximately 1000 times shorter than the x-ray pulses). This figure shows the relative intensity of the x-rays diffracted at a particular angle and time in a false color representation. In contrast to techniques for determining complicated protein structures, which require hundreds of diffraction peaks, here the scientists start with a well-known and relatively simple zinc-blende structure of the InSb crystal. They look for small changes, initiated by the laser absorption, in the dif-

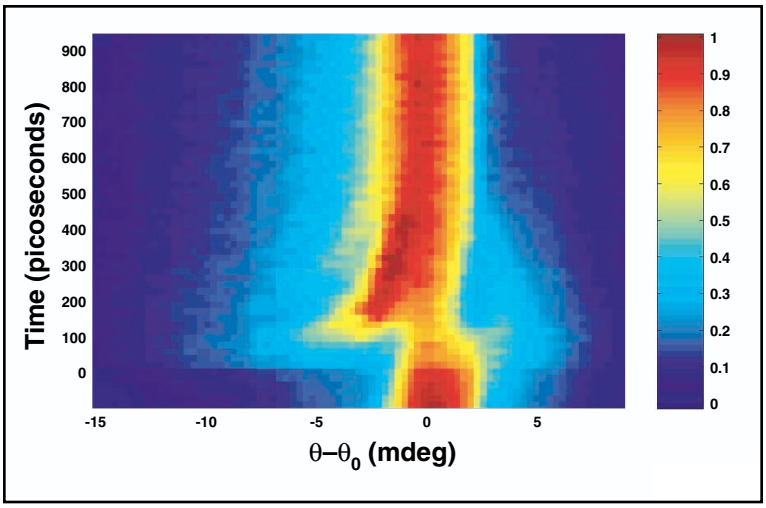

FIG. 2. Time-resolved diffraction pattern about the 111-peak of a rapidly heated InSb crystal.

fraction condition about a single peak as they vary both the time delay and angle of incidence of the well-defined incident x-ray beam. Each value of the angle and time shown in Fig. 2 was the result of approximately 1000 exposures. To obtain an accurate picture requires extremely precise control over the experimental parameters during more than an hour of data collection.

When the short laser pulse is absorbed by the material, it differentially heats a thin surface layer of the crystal, which leads to an expansion of the lattice near the surface. This launches a short acoustic pulse, which comprises a region of lattice compression followed by lattice expansion [2]. This acoustic pulse is only a few tens of picoseconds in duration and comprises frequencies over one million times higher than those audible by the human ear. As the acoustic pulse propagates into the crystal, the heated surface region relaxes back to its equillibrium condition before the arrival of the next laser pulse. By comparing the diffraction pattern in Fig. 2 to model calculations, the researchers have determined that the standard thermoelastic model for acoustic pulse generation underestimated the magnitude of the disturbance. These experiments are at the forefront of time-resolved experiments at synchrotrons. Despite the already impressive temporal resolution in these experiments, to better understand the physical mechanism requires even better temporal resolution.

Producing even shorter x-ray pulses from the synchrotron is a major motivation in this research. This will open the door to femtosecond studies at current third-generation synchrotrons and build the foundation for ultrafast x-ray science at femtosecond free-electron and other linear-accelerator-based sources of the future.

Principal publication: "Probing Impulsive Strain Propagation with X-ray Pulses," Phys. Rev. Lett. **86**, 3072–3075 (2001).

References

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